

Atomic Mechanisms of Liquid Metal Spreading

MSD researchers Eduardo Saiz and Antoni Tomsia have elucidated the atomic mechanisms that underlie liquid metal spreading at high temperature. They found that the dynamics are fundamentally different from those involved in the better-understood case of room temperature liquids; they involve not viscosity, but processes occurring at the “triple line” where the liquid metal front, the solid substrate, and the gaseous atmosphere are all in contact.

The spreading of liquid metals plays a key role in many modern industrial processes such as brazing, thin film deposition, and microelectronic soldering. However, despite the apparent simplicity of the phenomenon, spreading of liquid metals at high temperatures has defied description and generalization. For example, the reported times for the spontaneous spreading of small metal drops on a surface range from milliseconds (as would be expected from a comparison with organic liquids of similar viscosity) to minutes and even longer. The challenge has been to link macroscopic measurements such as the dynamic contact angle or the speed of a moving liquid front to phenomena occurring at the microscopic and even atomic level in the vicinity of the triple solid-liquid-vapor junction. One of the roots of the problem is the difficulty in designing high-temperature experiments in which spreading can be recorded under precisely controlled conditions

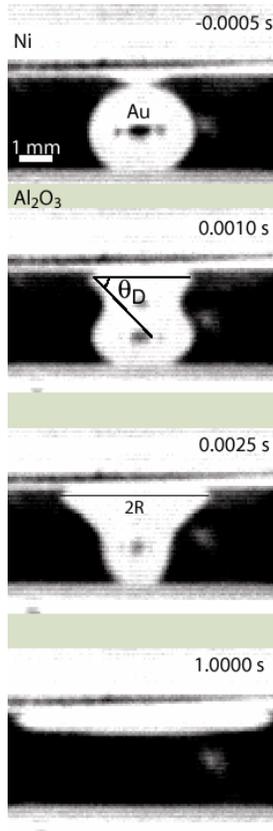
In a novel approach to this problem, the LBNL researchers manipulated small metal drops under controlled atmosphere and temperature and used high-speed photography to observe their spreading on solid substrates. Data from hundreds of tests with different metals and surfaces show that, counterintuitively, the spreading of the molten metal is not controlled by the viscous dissipation in the bulk liquid (i.e. by the liquid viscosity) as is commonly accepted in the analysis of the spreading of organic liquids at room temperature. Rather, liquid metal spreading is controlled at the atomic scale by the rate at which liquid atoms displace gaseous atoms at the triple line. This fundamental difference between the spreading of liquid metals and organic liquids of similar viscosity has its origin in the much stronger solid-liquid interactions in the metals in high-temperature systems.

During the study Saiz and Tomsia also observed for the first time the formation of “Marangoni films” at high temperatures. Marangoni films are caused by surface tension gradients and were first documented in organic liquids more than two hundreds years ago. Perhaps their best known manifestation is the “legs” or “tears” that form on the inside surface of a wine glass. The surface tension gradients that drive Marangoni films ahead of the liquid front are generated in this case by the evaporation of a highly volatile component in the liquid. Surprisingly they had not been observed in metal films until the LBNL researchers identified liquid metal systems in which variations in liquid composition that develop during the simultaneous spreading and interdiffusion generate the surface energy gradients that drive the formation of Marangoni films.

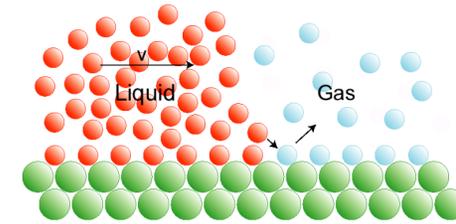
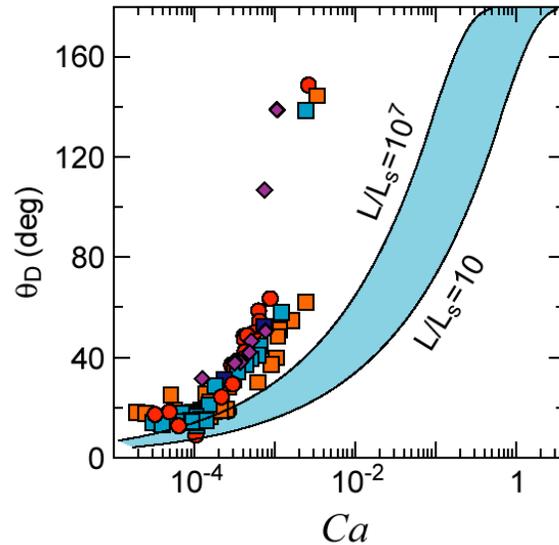
This work provides basic ideas and data that will guide the formulation of a unified theory of high-temperature spreading including the extremely important and still unresolved problem of the spreading of reactive liquids. The benefits will extend to the theoretical analysis of nonequilibrated interfaces and to the development of new technologies that require control of liquid metal flow at increasingly smaller dimensions.

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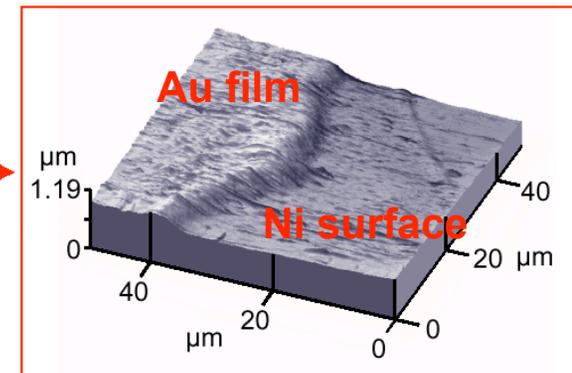
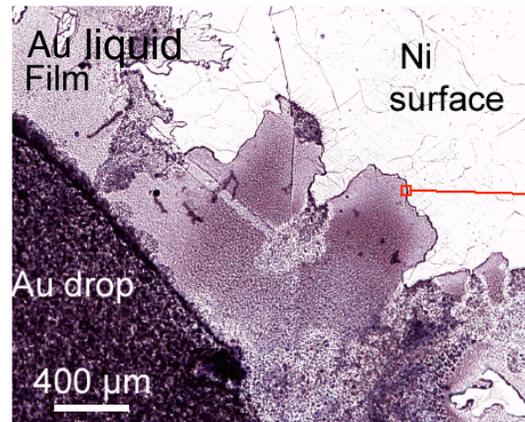
Eduardo Saiz and Antoni P. Tomsia, “Atomic dynamics and Marangoni films during liquid-metal spreading,” *Nature Materials* **3**, 903; December 2004



High speed photography is used to measure the contact angle (θ_D) and the speed of the advancing liquid front (Ca the rate of change of R) as small metal drops transfer from an inert substrate to a metallic substrate that approaches from above. Spreading occurs in just a few milliseconds.



The relationship between the dynamic contact angle (θ_D) and the normalized velocity of the liquid front (Ca , left,) of a variety of metal substrate pairs e.g., Au or Cu on nickel; (squares, circles, diamonds, left) and surfaces does not correspond to what would be expected if spreading were controlled solely by the viscous dissipation in the liquid (blue band). As depicted above, liquid metal spreading is instead controlled by the rate at which liquid atoms (red) displace gaseous atoms (blue) at the triple line where solid, liquid and gas meet.



In several metal systems (Au spreading on Ni is shown) a thin liquid film (light purple) moves ahead of the main wetting front (dark purple). Careful examination of the shape of the leading edge of the film (AFM image, right) show that it is a Marangoni film whose formation is driven by gradients in the liquid surface energy. This is the first time that Marangoni films have been observed in high-temperature spreading.